

# Lanthanide gallate perovskite-type substrates for epitaxial, high- $T_c$ superconducting $\text{Ba}_2\text{YCu}_3\text{O}_{7-\delta}$ films

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Previous studies had indicated promising use of lanthanide gallate perovskite-type substrates for the deposition of epitaxial, high- $T_c$  superconducting  $\text{Ba}_2\text{YCu}_3\text{O}_{7-\delta}$  (BYCO) films. They were also found to have moderate dielectric constants ( $\sim 25$  compared to  $\sim 277$  for  $\text{SrTiO}_3$ ). This study was undertaken to further explore the use of  $\text{LaGaO}_3$ ,  $\text{NdGaO}_3$ ,  $\text{SrTiO}_3$ ,  $\text{MgO}$ , and Y-stabilized  $\text{ZrO}_2$  substrates, prepared from single-crystal boules grown by several suppliers using the Czochralski

method. Films were prepared by cylindrical magnetron sputtering and laser ablation. Substrate evaluations included measurement of dielectric constant and loss, thermal expansion, and mechanical hardness and toughness. In addition to their moderate dielectric constants, they were found to have satisfactory mechanical properties, except for the twinning tendency of  $\text{LaGaO}_3$ . Lattice mismatch strains were calculated for orthorhombic BYCO films on a number of substrates.  $\text{NdGaO}_3$  was found to have the best lattice match with BYCO, and is now available twin-free.

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## Introduction

Recently, Sandstrom et al. [1] reported the first use of  $\text{LaGaO}_3$  substrates to produce good-quality epitaxial, high-

$T_c$  superconducting films. It was noted that the  $\text{LaGaO}_3$  perovskite-type crystal structure provides a good lattice match for epitaxial  $\text{Ba}_2\text{YCu}_3\text{O}_{7-\delta}$  (BYCO) film growth, and furthermore that the lanthanide gallates have lower dielectric constants ( $\epsilon \sim 25$ ) than  $\text{SrTiO}_3$  ( $\epsilon \sim 277$ ). Previously, in connection with the growth of magnetic ferrite garnet epitaxial films on nonmagnetic gallate garnet substrates, it had been observed that film and substrate lattice parameters had to match within 0.1% to obtain pseudomorphic dislocation-free films about one  $\mu\text{m}$  thick [2]. Otherwise, the films cracked when in tension, or slipped to form low-angle grain boundaries when in compression. One might reasonably expect BYCO films to suffer similar constraints.

Superconductivity is highly anisotropic in orthorhombic BYCO. The highest critical current densities  $J_c$  are realized in  $a$ - $b$ -plane single-crystal epitaxial films [3, 4]. However, even "epitaxial" polycrystalline films, which have (single-crystal) grains oriented with their  $c$ -axes all perpendicular to the film plane and with their  $a$ - and  $b$ -axes randomly oriented, can display relatively high  $J_c$  values [5, 6]. Both grain boundaries and intragranular twins in BYCO influence  $J_c$  [7, 8]. Twins form in BYCO crystals when they are cooled through the first-order tetragonal-to-orthorhombic phase transition at  $\sim 680^\circ\text{C}$  in an oxidizing atmosphere. A larger first-order phase transition has been observed at  $\sim 150^\circ\text{C}$  by dilatometry in Czochralski-grown  $\text{LaGaO}_3$  crystals [1, 9]; the high-temperature phase is rhombohedral [10].

With the laser ablation technique [11], Koren et al. [12] have deposited BYCO films on commonly used  $\text{SrTiO}_3$  substrates, and on  $\text{NdGaO}_3$  as well as  $\text{LaGaO}_3$  substrates.  $\text{NdGaO}_3$  was chosen because it has an even better lattice match with BYCO than does  $\text{LaGaO}_3$ . The  $J_c$  values obtained for films deposited onto  $\text{NdGaO}_3$  were only slightly less than those deposited onto  $\text{SrTiO}_3$ ; the use of  $\text{LaGaO}_3$  resulted in lower  $J_c$  values.

More recently, Miyazawa [13] has used a mechanical profilometer to measure the degradation in the surface morphology of  $\text{LaGaO}_3$  that is heat-cycled through its  $150^\circ\text{C}$  phase transition. He suggested that the relatively low  $J_c$  for BYCO films on  $\text{LaGaO}_3$  results from the associated surface roughening.

While Simon et al. [14] have shown that  $\text{LaAlO}_3$  single crystals can also be used as substrates for BYCO films, Feenstra et al. [15] found that the use of such substrates typically results in a reduction of  $T_c$  by 6–8 K, compared to that which results when  $\text{SrTiO}_3$ ,  $\text{KTaO}_3$ , and  $\text{LaGaO}_3$  are used; their films were produced by co-evaporation of Y, Cu, and  $\text{BaF}_2$  followed by annealing at  $850^\circ\text{C}$  in wet  $\text{O}_2$ . Although this film preparation method can produce more film reactivity with substrates than the lower-temperature laser ablation method, it should be noted that whatever deposition method is used,  $\text{LaAlO}_3$  is rhombohedral rather

than orthorhombic [10] and contains microtwins. Nevertheless, other investigators [16] using  $\text{LaAlO}_3$  have reported favorable results, and have indicated a preference for it.

This paper reports experimental data for several types of single-crystal boules and substrates prepared to our specifications by several suppliers, and for BYCO films deposited onto the substrates in our laboratory, using laser ablation sputtering [11, 12] and cylindrical magnetron sputtering [17]. Taking into account previous, relevant crystallographic and phase equilibria investigations [9, 18–29], we examined the following substrate properties: (1) fracture and deformation behavior (examined by using mechanical indentation techniques); (2) the effects of film processing and use conditions (superconducting device configurations formed on the substrates would need to survive repeated stress-inducing heating and cooling cycles); and (3) dielectric constants and thermal expansion coefficients.

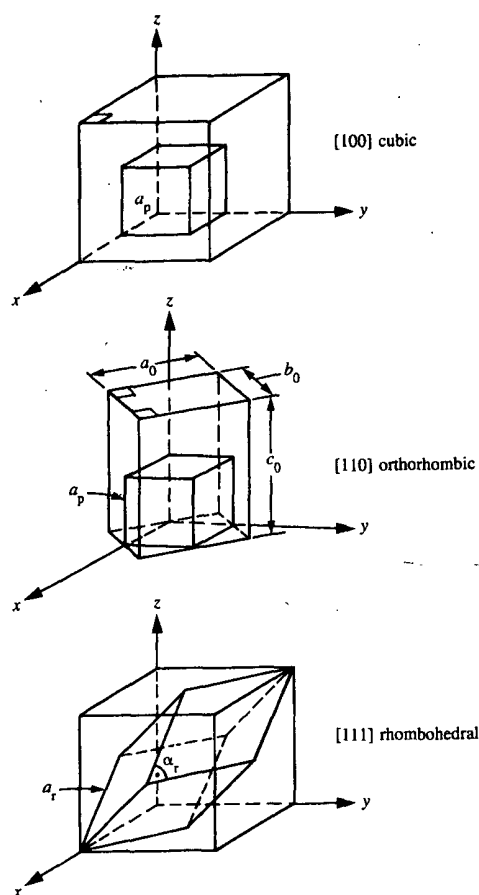
### Crystallography and crystal chemistry

The primitive perovskite structure is cubic. The designation "perovskite" originates from the mineral  $\text{CaTiO}_3$ , which has a unit cell of Ti atoms located at the eight corners of a cube, O atoms centered on the twelve edges of the cube, and one Ca atom at the center of the cube. In  $\text{LaGaO}_3$ , for example, La replaces the Ca, and Ga replaces the Ti. All of the larger lanthanides can replace the Ca at the cube center, and, for example, Cu or Al can replace the Ti.

The unit cell of BYCO is composed of two primitive " $\text{BaCuO}_{3-x}$ " cells having a primitive cell of " $\text{YCuO}_{3-y}$ " sandwiched between them. Thus, the unit cell of BYCO is tripled in the  $c$  direction; its lattice parameters are  $a = 3.827 \text{ \AA}$ ,  $b = 3.877 \text{ \AA}$ , and  $c = 11.708 \text{ \AA}$  [30]. Because the cell has a [100]-type orthorhombic distortion,  $a \neq b$ .

The lanthanide gallate substrates of interest are those that contain the larger rare earths and La; the structure of these gallates is orthorhombic (see [110] in Figure 1) at room temperature and rhombohedral ([111] in Figure 1) near their melting (growth) temperatures. Note that BYCO has a [100] distortion from the primitive cubic perovskite-type structure, whereas the orthorhombic lanthanide gallate substrates have a [110] distortion; hence their  $a$ - and  $b$ -axes are  $\sqrt{2}$  larger than and are rotated about  $45^\circ$  from the primitive cubic structure values. As Geller [10] has observed, the orthorhombic gallates actually have a pseudocubic cell distorted into the form of a rhombus with parameters  $a = b \approx 3.9$  and nonorthogonal ( $\alpha \neq 90^\circ$ )  $a$ - and  $b$ -axes.

This notwithstanding, the simplest way of lattice-matching the two related, but different,  $\text{ABO}_3$  perovskite-type orthorhombic space groups is to consider how unit cell areas must meet at the film-substrate interface.



**Figure 1**

Perovskite-type crystal systems: [100] cubic, [110] orthorhombic, and [111] rhombohedral.

Although the substrate and film lattices are distorted differently from the simple pseudocubic cell, a direct comparison of their  $ab$  planes can be made by dividing the  $a_0$  and  $b_0$  of the substrate by  $\sqrt{2}$ , because the film unit cell contains only half as many atoms in the plane. This normalization gives a test for the condition in which the same number of atoms per unit area exist for both sides of the film-substrate interface (a low-strain condition). Thus, it is used in Table 3 (shown later) to estimate misfit strains for BYCO films on gallate substrates.

### Lanthanide gallate perovskites

In 1957, Roth [31] published a comprehensive paper on the crystallography of perovskite and other  $ABO_3$ -type compounds. The radii, valences, and polarizabilities of the

A and B ions are important in structural relationships. Geller [10] studied perovskite-like compounds with A = the lanthanides and B = Sc, V, Al, Ga, Cr, showing many to be isostructural with  $GdFeO_3$ . He later [20] grew Czochralski crystals of  $NdGaO_3$  to serve as substrates for ferrite films. Furthermore, he found the orthorhombic-rhombohedral-cubic phase transition sequence for these gallate perovskite compounds. Geller et al. [19] measured the orthorhombic lattice parameters for the lanthanide gallate series from La to Er, which are  $5.52 \geq a_0 \geq 5.24 \text{ \AA}$ ,  $5.49 \leq b_0 \leq 5.52 \text{ \AA}$  (but with a maximum of 5.54 for Gd), and  $7.77 \geq c_0 \geq 7.53 \text{ \AA}$ , respectively. They found  $a_0 \approx b_0$  for  $CeGaO_3$ , which indicates that it is pseudotetragonal. The heavier (smaller) rare-earth gallates were found not to form stable perovskites. Marezio et al. [21] grew La, Pr, and Nd gallate crystals from melts fluxed with lead borate, and measured lattice parameters essentially in agreement with those of Geller and his co-workers.

Recently, the Czochralski method has been used by Belt and Uhrin [32] and by Berkstresser et al. [33] to produce >25-mm-diameter single-crystal boules of  $LaGaO_3$  and  $NdGaO_3$ , as well as  $LaAlO_3$ . The boules were oriented, sliced, and polished into substrate wafers for epitaxial film studies in their laboratory and ours.

### Thermal expansion behavior

The present trend in the deposition of BYCO films is to maintain substrate temperature in the range 650–800°C during deposition. Because the thermal expansion mismatch strains introduced during cooling to room temperature and subsequent annealing runs can lead to microcracking or the generation of defects in the films, resulting in degradation of their critical current densities, thermal expansion behavior and potentially disruptive substrate phase transitions that may occur in the applicable temperature range should be taken into consideration.

Figure 2 shows the thermal expansion behavior of three different lanthanide gallate single crystals, examined from 25–900°C using a Dilatronic Model 1200C dilatometer (Theta Industries, Port Washington, NY). Measurements were made along the [001] growth directions. The  $LaGaO_3$  single crystals were heavily twinned and showed a sharp (first-order) phase transition at 147°C. The dilatation associated with this transition was about 0.3%. The coefficient of thermal expansion (CTE) above the transition was determined to be  $10.3 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$ , which differs only slightly from that found for BYCO ( $12.0 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$ ). However, the presence of a disruptive transition at 147°C makes  $LaGaO_3$  a less desirable substrate.

We have also studied the effects of substitutions on the La site in the crystal structure on the transition temperature and CTE; Nd substitution on a third of all La sites shifted the transition to about 710°C. The transition

was no longer sharp (710–715°C) and exhibited some hysteresis, and the CTE dropped to  $8.72 \times 10^{-6} \text{°C}^{-1}$ . Finally, pure  $\text{NdGaO}_3$ , in which Nd atoms replace La atoms, showed no transition up to the highest measured temperature (900°C). The  $\text{NdGaO}_3$  crystals were found to be almost twin-free, and the CTE lower ( $8.02 \times 10^{-6} \text{°C}^{-1}$ ).

The overall dilatation of the three lanthanide gallates examined differed by only a few percent at 900°C. On the basis of the absence of any transitions in the temperature range of BYCO thin-film processing,  $\text{NdGaO}_3$  was found to be the best of the three. Furthermore, the CTE mismatch is slight. Thus, internal strains that develop after film growth upon cooling to room temperature should be minimal.

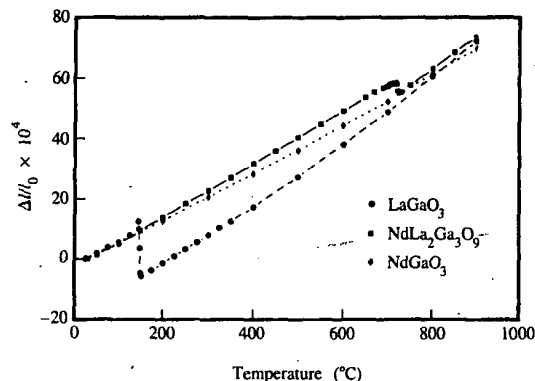
The  $\text{La}_2\text{NdGa}_3\text{O}_9$  crystals were grown to ascertain whether an ordered and tripled perovskite cell isostructural with  $\text{Ba}_2\text{YCu}_3\text{O}_{7-x}$  would form; however, no tripled cell was detected (using X-ray diffraction). Kokta and Grasso [34], for example, found La and Lu atoms ordered in  $\text{La}_3\text{Lu}_2\text{Ga}_3\text{O}_{12}$  garnets. Because the atomic radii of La and Lu differ by so much more than do those of La and Nd, there is a greater tendency toward the formation of ordered structures.

### Hardness and toughness

The fracture and deformation properties of several types of substrates were examined by using indentation techniques. Hardness and toughness values obtained are shown in Table 1. To obtain the values, a Vickers diamond pyramid was loaded onto the polished surfaces of the indicated types of single-crystal substrates at peak loads between 0.1 and 10 N. The resulting contact impression dimensions and radial crack lengths were measured by optical microscopy. The tendency for the cracks to follow cleavage planes in the crystals was noted in all cases, and the indentations aligned to yield cracks propagating colinearly with the contact impression diagonals.

The impression diagonal dimensions varied with contact load with a power law dependence of 1/2, indicating constant hardness. The crack lengths varied with contact load with a power law dependence of 2/3, indicating constant toughness. Although toughness values are model-dependent, they could be estimated to an accuracy of  $\pm 20\%$  [35].

The low toughness of  $\text{LaGaO}_3$  may not relate to the twinning tendency it exhibits.  $\text{NdGaO}_3$ , which does not twin so badly because it has crystallographic transition temperature greater than 1000°C, displayed the same toughness as  $\text{LaGaO}_3$ . It is interesting to note that "ferroics," which are ferroelectric solid solutions that have smeared- instead of sharp-transition temperatures, have finer-scale domain structures.  $\text{La}_{0.9}\text{Gd}_{0.1}\text{GaO}_3$  crystals exhibit finer-scale twin spacings.  $\text{LaAlO}_3$  is rhombohedral at room temperature, and it has a twinned structure that



**Figure 2**

Thermal expansion of crystals of  $\text{LaGaO}_3$ ,  $\text{La}_2\text{NdGa}_3\text{O}_9$ , and  $\text{NdGaO}_3$ .

**Table 1** Mechanical properties of substrates.

	Hardness (GPa)	Toughness ( $\text{MPa} \cdot \text{m}^{1/2}$ )
$\text{LaGaO}_3$	9.4	0.7
$\text{NdGaO}_3$	9.4	0.7
$\text{SrTiO}_3$	5.0	1.0
$\text{MgO}$	7.6	1.3
$\text{Y-ZrO}_2$	17.8	1.2

results from a  $\sim 470^\circ\text{C}$  phase transition [9], above which the structure is cubic up to the melting point [10]. Thus, the phase transitions producing twins in  $\text{LaAlO}_3$  and  $\text{LaGaO}_3$  are different. Indeed,  $\text{LaGaO}_3$  also may have a rhombohedral-to-cubic phase transition near its melting point.

Although the hardness of  $\text{LaGaO}_3$  is greater than that of  $\text{SrTiO}_3$ , its toughness is lower; hence, neither appears to be clearly superior. Generally, the gallate perovskites show properties comparable to those of other oxides. On the other hand,  $\text{Y-ZrO}_2$  seems to be the best of the four with regard to these mechanical properties.

### Dielectric properties

Dielectric measurements were made on  $\sim 1\text{-mm}$ -thick disks with their parallel flat surfaces coated with a flexible silver paint (Englehard Co., Newark, NJ). Samples were vacuum dried at  $110^\circ\text{C}$  for 1 h before measurement. Capacitances were measured using a 4275A Multi-Frequency LCR Meter (Hewlett Packard Co., Cupertino, CA), at 0.1 V and frequencies

**Table 2** Dielectric constant ( $\epsilon$ ) and loss ( $\tan \delta$ ) values at 1 MHz, measured at room temperature. The indicated (hkl) values are for pseudocubic perovskite.

	(hkl)	$\epsilon$	$\tan \delta$	Supplier
LaAlO <sub>3</sub>	(100)	23	0.007	AT&T
LaGaO <sub>3</sub>	(001)	25.0*	0.0018	Airtron, AT&T
NdGaO <sub>3</sub>	(001)	20.2	0.002	Airtron, AT&T
Y-ZrO <sub>2</sub>	(100)	27.5	0.0054	Ceres
MgO	(100)	9.62	0.0091	Atomergic
SrTiO <sub>3</sub>	(100)	277	0.0125	Commercial
SrTiO <sub>3</sub>	(110)	170	0.0130	Commercial
Nd-YAlO <sub>3</sub>	(001)	16.0	0.0010	Airtron

\*From Reference [1].

Airtron: Airtron Div. Litton, Morris Plains, NJ

AT&T: AT&T Nassau Metals, Irmo, SC

Ceres: Ceres Corp., No. Billerica, MA

Commercial: Commercial Crystals Lab, Naples, FL

Atomergic: Atomergic Chemetals, Farmingdale, NY

of 10 kHz, 100 kHz, 1 MHz, and 10 MHz. Samples were shielded by an aluminum cage during measurement.

The results obtained are shown in Table 2. As can be seen, the  $\epsilon$  values of the perovskite aluminates and gallates are lower by an order of magnitude than that of SrTiO<sub>3</sub>, and therefore should be superior with regard to signal propagation. Their  $\tan \delta$  values are also lower. Use of the larger lanthanide ions La and Nd results in higher  $\epsilon$  values than when use is made of smaller ions such as Y. Although the value of  $\epsilon$  is lowest for the MgO, that material is less desirable from the standpoint of lattice mismatch with the cuprates (discussed later).

## Epitaxial films

In order to fabricate useful, high- $T_c$  superconducting devices, it would be desirable to have the ability to produce single-crystal, single-domain, untwinned epitaxial films that could be patterned. It would be preferable if such films could be grown on the surface of bulk single crystals having an appropriate crystallographic plane matching the arrangement and the spacing of atoms in the desired film. Most often the substrate crystal has a different chemical composition, so that it is passive and does not interfere with device phenomena. Ideal film-substrate combinations result in "pseudomorphic" films with perfect physical matching of the substrate.

Although this has not been achieved for BYCO films, such matching should be possible if the substrate were orthorhombic from room temperature to film processing temperatures, which generally exceed that of the BYCO orthorhombic-to-tetragonal phase transition. This condition should facilitate formation of the desired superconducting orthorhombic phase in cases in which the substrate does not form twins, and thus should make it possible to produce a single-domain (untwinned) orthorhombic epitaxial BYCO film. NdGaO<sub>3</sub> appears to be the best substrate candidate for this purpose.

## Epitaxial strains

Matthews and Klokholm [36] have defined the elastic (misfit) strain in a "pseudomorphic" film in perfect registration at the interface with its substrate at room temperature as

**Table 3** Epitaxial misfit and thermal strains for BYCO films on various substrates.

	Lattice parameters			Mismatch			Thermal	
	a (Å)	b (Å)	c (Å)	$\epsilon_a$ (ppt)	$\epsilon_b$ (ppt)	$\epsilon_c$ (ppt)	CTE (ppm/°C)	$\epsilon_T$ (ppt)
YAlO <sub>3</sub>	3.660	3.678	3.753	-45	-54	-40		
LaAlO <sub>3</sub>	3.793			-9	-22	-30		
YGaO <sub>3</sub>	3.717	3.915	3.766	-29	10	-36		
LaGaO <sub>3</sub>	3.885	3.903	3.885	15	7	-5	10.3	1.0
LNdGaO <sub>3</sub>	3.864	3.879	3.870*	10	1	-9	8.7	2.0
NdGaO <sub>3</sub>	3.837	3.889	3.853	3	3	-13		
	3.829	3.878	3.845*	1	-2	-15	8.0	2.4
LaSrGaO <sub>4</sub> <sup>†</sup>	3.843	3.843	4.227	4	-9	-77		
SrTiO <sub>3</sub>	3.905			20	7	1	10.4	1.1
YZrO <sub>2</sub>	3.607			-61	-75	-82	9.2	1.7
MgO	4.213			92	80	74	12.6	-0.4
BYCO film	3.827	3.877	3.903	0	0	0	12.0 <sup>‡</sup>	0.0

\*Grown by Czochralski method

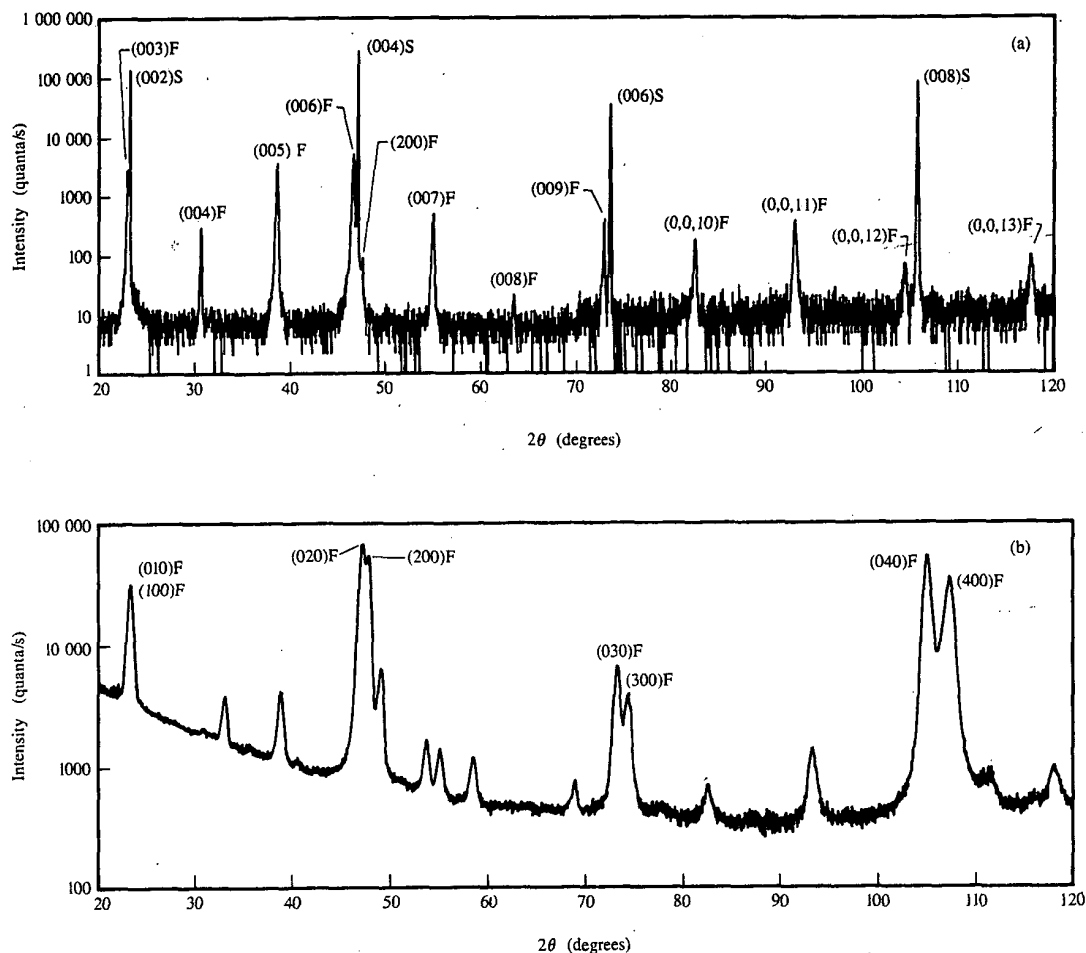
†Reported by B. H. T. Chai

‡Average for a ceramic

LNdGaO<sub>3</sub> = La<sub>0.7</sub>Nd<sub>0.3</sub>GaO<sub>3</sub>

ppt = parts per thousand

ppm = parts per million



**Figure 3**

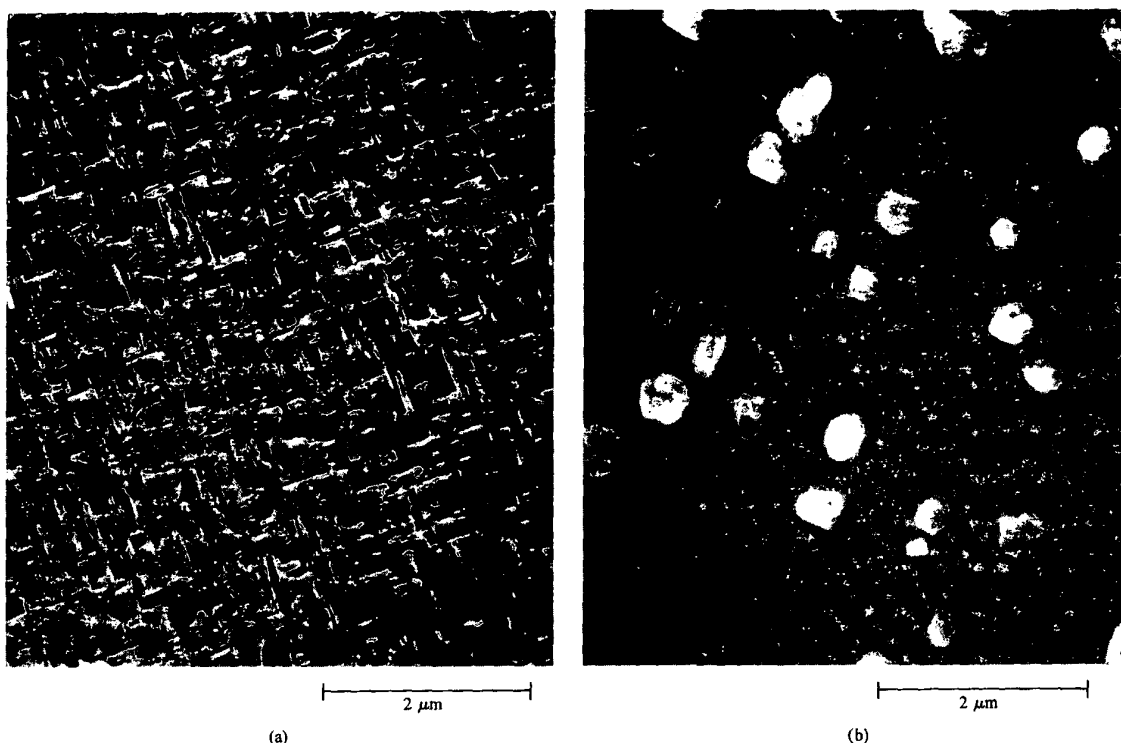
(a) A  $\theta$ - $2\theta$  double-crystal X-ray diffraction pattern from a  $\text{Ba}_2\text{YCu}_3\text{O}_{7-\delta}$  film deposited onto  $\text{NdGaO}_3$  [001], recorded in the symmetric Bragg mode. S and F designate substrate and film reflections, respectively. (b) A grazing-incidence X-ray diffraction pattern from the same sample.

$$\epsilon_a = \frac{a_{\text{film}} - a_{\text{substr}}}{a_{\text{film}}},$$

where  $a_{\text{film}}$  and  $a_{\text{substr}}$  are the lattice parameters of the film and substrate. They showed the importance of the associated strain energy in determining how much mismatch films can sustain before undergoing physical deterioration. Thinner films would thus be expected to tolerate a greater mismatch. For example, they predicted that a 3-nm-thick garnet film should be able to accommodate a 1% mismatch, whereas a 1- $\mu\text{m}$ -thick film should only be able to accommodate a 0.02% mismatch.

Table 3 compares lattice parameters of BYCO with normalized values of some potential substrates. The normalized values were calculated for a simple perovskite unit cell. This is an approximation, since the space groups are usually not the same. Specifically, as mentioned earlier, the BYCO is distorted in the [100] direction, while the gallates are distorted along the [110] direction in the case of orthorhombic distortion, and along the [111] direction in the case of rhombohedral distortion.

Thermal expansion must be taken into account if the lattice parameters have been measured at room



**Figure 4**

Scanning electron micrographs of the surfaces of  $\text{Ba}_2\text{YCu}_3\text{O}_{7-\delta}$  films deposited (a) by sputtering; (b) by laser ablation.

temperature and film growth is to occur at an elevated temperature (as discussed earlier). The thermal expansion mismatch is

$$\epsilon_T = 600^\circ\text{C} \times (\text{CTE}_{\text{film}} - \text{CTE}_{\text{substr}}).$$

The magnitude of the strain  $\epsilon_T$  (due to thermal expansion) is less than that of  $\epsilon_a$  (due to lattice mismatch). Of all the substrate types listed,  $\text{NdGaO}_3$  produces the lowest  $\epsilon_a$ . Since it can now be grown twin-free [33], the possibility exists of using it to produce twin-free epitaxial films of BYCO.

#### X-ray diffraction of films

For structural characterization of the BYCO film deposited by laser ablation, X-ray diffraction was performed in the grazing incidence (GID) and symmetric Bragg modes, for which the probing directions are parallel and perpendicular to the interface, respectively [37]. The symmetric Bragg-mode pattern from a BYCO film grown on a  $\text{NdGaO}_3$  substrate is shown in Figure 3(a), where peaks from the film and substrate are designated by an F or S, respectively. Those from the film were its (001) reflections, indicating that it was preferentially oriented with its *c*-axis normal to the substrate. Figure 3(b) shows the GID pattern

that was obtained, with primarily the (h00) and (0k0) reflections present, suggesting that the film was epitaxial in the plane of the substrate, with two domains. The X-ray results suggest that the film was a twinned single crystal in the *a*-*b* plane, having its *c*-axis perpendicular to the surface of the substrate.

#### Scanning electron microscopy films

Figure 4 shows scanning electron micrographs of two BYCO films, prepared by different deposition methods, on  $\text{NdGaO}_3$ [001] substrates. Because of the recent success by Xi et al. [17] in using cylindrical magnetron sputtering, we chose to make use of that method to grow our epitaxial BYCO films. The sputtered film depicted in (a) showed a unique growth morphology compared to that depicted in (b), fabricated by laser ablation. High-resolution secondary-electron images of the film in (a) indicated the existence of both *a*-axis (perpendicular to the substrate plane) and *c*-axis epitaxy. Specifically, the film of (a) appeared to be oriented mostly with *c*-axis portions in the plane of the film (leading to a lower critical current density), in accordance with Bragg X-ray diffraction data (not shown). A block-like surface texture dominated; the blocks were highly oriented  $90^\circ$ , 200–500-nm block-like

regions that were interpreted as being oblong as a result of the difference in crystal growth velocities along the *a*- or *b*- and the *c*-axis. (Growth along the [001] direction is significantly slower [38].)

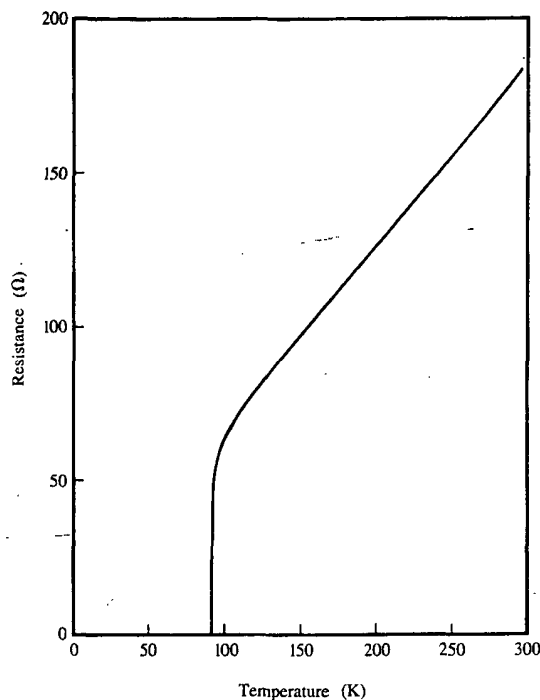
By comparison, the film depicted in (b) contained large, continuous flat-plate regions and was oriented with its *c*-axis normal to the plane of the film (characteristic of a film capable of supporting a high critical current density). The "boulders," especially in the film of (b), were probably vapor-transported aggregates rather than being a result of coalescence fed by surface diffusion. R. Ramesh et al. [39] have conducted an HREM lattice image analysis of sections of BYCO films grown on MgO, SrTiO<sub>3</sub>, LaAlO<sub>3</sub>, and other substrates. They have found that although MgO has essentially no lattice match with BYCO, it nucleates films having a coincident site lattice and their *c*-axes normal to the substrate plane. There are multiples of [110] BYCO and [100] MgO spacings that promote ordering. Nevertheless, it is reported [39] that BYCO films on MgO display a critical current density which is an order of magnitude smaller than that of BYCO films on SrTiO<sub>3</sub>. Growth in the BYCO [001] *c*-axis direction is significantly slower than in the [010] or [100] direction. Thus, when films having their *c*-axes normal to the substrate surface grow, they tend to follow a lateral step/ledge motion (Frank-van der Merwe mechanism) which often results in *c*-axis stacking faults. Interestingly, at low growth temperatures (700°C), normal growth in the direction of the *a*-axis (the rapid-growth direction) occurs, and there is atomic registration in columns perpendicular to the substrate. At higher growth temperatures and in film layers nearest to the substrates, the *c*-axis is normal to the substrate plane.

### Film resistivity and critical current

A typical plot of resistance versus temperature for a 200-nm-thick BYCO film deposited by laser ablation onto a NdGaO<sub>3</sub> substrate is shown in Figure 5. The film exhibited good metallic behavior in the normal state, with its resistivity extrapolating to zero with temperature. The superconducting transition upon cooling was sharp, with onset and zero-resistance temperatures at 92.5 K and 90.7 K, respectively. The critical current density of the film in zero magnetic field was measured using a patterned 25-μm-wide, 200-μm-long microstrip of the film, using a threshold criterion of 1 μV/mm. The critical current density was found to be  $1 \times 10^6$  A/cm<sup>2</sup> at 86 K, and  $2 \times 10^6$  A/cm<sup>2</sup> at 82 K, values as high as those measured for the best BYCO films grown on SrTiO<sub>3</sub> substrates.

### Summary

Among the single-crystal lanthanide substrates examined in this study, NdGaO<sub>3</sub> was found to be the most suitable of the group for the deposition of epitaxial Ba<sub>2</sub>YCu<sub>3</sub>O<sub>7-δ</sub>



**Figure 5**

Typical plot of resistance versus temperature for a Ba<sub>2</sub>YCu<sub>3</sub>O<sub>7-δ</sub> film deposited onto a NdGaO<sub>3</sub> substrate by laser ablation, showing superconducting-to-normal transition.

superconducting films, primarily because of the absence of any substrate twin-generating transitions in the temperature range of interest. Also, calculations of substrate-film mismatch strains indicated that NdGaO<sub>3</sub> had the best lattice match of the group—better than that of LaGaO<sub>3</sub>. The mechanical hardness and toughness of the NdGaO<sub>3</sub> substrates were found to be similar to those of commonly used SrTiO<sub>3</sub> substrates. Measurements of their dielectric properties indicated low loss and a dielectric constant that was an order of magnitude less than that of SrTiO<sub>3</sub> substrates. Given the attractive possibility of forming twin-free NdGaO<sub>3</sub>, it would therefore appear to be worthwhile to attempt to produce twin-free Ba<sub>2</sub>YCu<sub>3</sub>O<sub>7-δ</sub> films on such a substrate, thus potentially leading to a substrate-film combination having favorable mechanical, dielectric, and superconducting properties.

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